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ELECTRICAL POTENTIALS IN FREEZING SOLUTIONS AND THEIR EFFECT ON MIGRATION

R. I. Korkina

Cold Regions Research and Engineering Laboratory

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R.I. Korkina





CORPS OF ENGINEERS, U.S. ARMY
COLD REGIONS RESEARCH AND ENGINEERING LABORATORY
HANOVER, NEW HAMPSHIRE

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Electrical potential originating			
water, weak salt solutions, and c	lay suspensions v	was studied experimentally	
in relation to moisture migration	in soil during i	freezing. The experimental	
procedure consisted in keeping the	a sample-filled i	flasks, insulated on all	
sides but the bottom, under define vating the variation of the magnit	ice minus tempera	sture conditions and obser-	
in and the table of the magnit	and ståll OI	potential in time. Curves	

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Unclassified SECURITY CLASSIFICATION OF THIS PAGE(When Data Entered) relating these variations to the distance between the measuring electrode and the cooling surface, and to cooling time were plotted from the reading obtained. Interpretation of the results indicated that a change in water structure preceeding its freezing was the cause of origin of electrical potential and that this effect on water migration in freezing soil is dependent on the concentrations of salts dissolved in the soil moisture, the mineralogical composition of colloidal particles suspended in the solution, and the composition of exchange cations on the surface of these particles.

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Table 1

ELECTRICAL POTENTIALS IN FREEZING SOLUTIONS AND THEIR EFFECT ON MIGRATION

Moscow AKADEMIYA NAUK SSSR. SIBIRSKOYE OTDELENIYE. INSTITUT MERZLOTOVEDENIYA. PROTSESSY TEPLO I MASSOOBMENA V MERZLYKH GORNYKH FORODAKH in Russian 1965, pp 56-65

[Article by R. I. Korkina]

[Text] The electric effect phenomenon arising in a one-sided freezing of diluted aqueous solutions is most fully demonstrated in the paper of E. Y. Workman and S. E. Reynolds (1950). They established that in one-sided freezing of water containing a small quantity (10⁻³ to 10⁻⁶N) of substances, capable of dissociation into ions in aqueous solutions there originates a comparatively large voltage difference on the waterice boundary. Maximal voltage values were obtained for a number of substances. Some of these values are given in Table 1.

Electric characteristics of some solutions (E. Y. Workman, S. E. Reynolds, 1950)

		2)	¥	<u></u>		٠	2 i .	4)
,	Вещество	Мансималь- ная разность потенциялов на грянице растаор — лед	Конпент- рации растнора, М	Удельное сопрозив- ление, ом/сж	Ј Вещество	Мансимадъвал разкитъ по- тенциалов на границе рас- твор — лед	Концент- рапия раствора. N	Удельное сопротив- ление, ом/см
5)	Н ₁ О би-	~-60		2,5-104	KCI	1 .	200×10-4	
•,	CaCl ₂ NaCl	+ 14 + 30	 100 × 10 *	(s. \$0#	NH₄GI NH₄OH	1 05 232	70×10°° 30×10°°	-

Key: 1—substance; 2—maximum voltage difference on the solution-ice boundary; 3—concentration of solution N; 4—specific resistance, ohm/cm; 5—bidistillate.

E. Y. Workman and S. E. Reynolds consider that the electric effect appears at the start of the freezing of the liquid and disappears when the freezing is completed. A potential barrier is formed at the surface of the boundary between the water and ice, and is preserved during the process of freezing the liquid phase. The voltage difference does not depend on the speed of freezing, but on internal current leakage which reduces the observed voltage for small speeds of freezing. At large freezing speeds, the improperly oriented ice crystals also reduce the value of the originating voltage difference. The electrical effect, in this case, is small or there is no electrical effect. The sign and value of the obtained voltage difference is a function of the nature and concentration of the substances in the squeous solution. A change of the voltage polarity in the solution with respect to the ice is explained by the selective insertion of ions of one polarity into the ice.

Similar tests were made by F. I. Bayadina (1960). She measured the voltage difference when freezing distilled water with a conductivity of 5 x 10⁻⁶ or 6 x 10⁻⁶ ohm. cm. The observed values of voltage difference varied within wide limits from 5-6 to 90-100 volts, with prevailing values being below 50 volts. F. I. Bayadina considers that, along with adsorption by the ice of ions of one polarity, there are other factors that also affect the origination and values of voltage such as crystallization processes, the difference in speed of freezing water in various parts of the dish, and processes of change in the structures of water and ice when their temperatures change. She established that the more transparent the ice is, i.e., the fewer air bubbles it contains, the greater the maximum value of the voltage is.

The contact voltage difference between water and ice was measured by V. I. Arabadzhi (1948, 1956). His tests indicated a voltage difference between water and ice varying within limits of 170 ± 25 millivolts.

The paper by Yumikis (1958) mentions that an electrical voltage originates in frozen ground systems. The value of the electromotive force depends, in this case, on the concentration of the electrolyte in the given ground. The author determined in the laboratory that in frozen ground systems consisting of Danellen ground (material carried out by glaciers), there originate electromotive forces from 40 to 120 millivolts. His paper suggests that, depending upon the properties of the porous material and several other factors still not fully explained, the originating voltage may facilitate the flow of moisture in the same direction as the action of the thermal voltage or in the opposite direction.

This article cites the results of tests on measuring voltages originating for the one-sided freezing of a bidistillate of weak solutions of salts and suspensions which make it possible to clarify certain patterns of

voltage change depending upon a number of factors. These investigations were made in order to study the effect of originating voltages on the migration of moisture when freezing.

The freezing of water, water solutions and suspensions was done in a cold room equipped with automatic temperature regulation. The measuring vessels were plexiglass cylinders (internal diameter 68 mm, 80 mm high) with a cover and bottom wade of various materials: stainless steel, red copper and aluminum. A platinum plate on the cover served as an electrode for measuring voltages originating in freezing solutions and for determining the polarity of the voltage.

To create conditions for freezing on one side only, the solution in the cylinder was frozen from below from the side of the metallic bottom. The cylinder was placed in a brass container 300 mm in diameter and 330 mm high. The cylinder was insulated on top and on the sides with wool and from rubber. The electrodes were connected to the device by a shielded conductor: the bottom of the cylinder-to the shielding sleeving, platinum electrode -- to the central conductor. To eliminate the possibility of induced interference from outside sources on the measured voltages originating when the solution freezes, the brass container, the shielding sleeve and the meter were grounded.

The voltage meter was a tube voltmeter type 187a that can measure DC voltages from 0.1 to 300 volts with high internal resistance (50 megonms).

A negative temperature corresponding to the freezing temperature of the solution was established in the room before the start of the test. A solution was poured into the cylinder to a certain mark (corresponding to a volume of 160 millileters) and it was placed in a brass container with thermal insulation. The brass container was then placed in the cold room and the time of the start of freezing was noted. Observations of changing the value and polarity of voltage with time were started from this moment. The tests were made primarily in a plexiglass cylinder with a red copper bottom. The tests were made also with weak solutions of NaCl, NH4, Cl, NH4OH. The following patterns of voltage changes were established as a result of the tests.

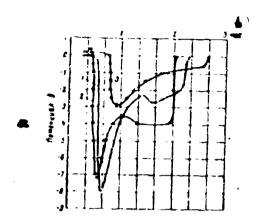


Fig. 1. Voltage change in the bidistillate depending upon the cooling time and the material of the cooled surface (cooling time $T = -30^\circ$; distance between electrodes h = 10 mm);

1--cooling through brass-red copper; 2--through brass-aluminum;

3--through brass-stainless steel.

Key: a--voltage, volts; b--hours.

The maximum voltage originating for one-sided freezing in a bidistillate and for a weak solution of NH₄Cl depends on the intensity of heat loss through the cooling surface and on the cooling temperature (Table 2). Changes in the voltage, depending upon the time of cooling and the material of the cooled surface, are shown on curves (Fig. 1)

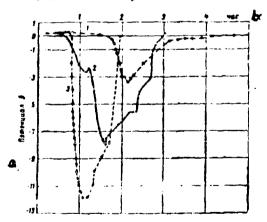


Fig. 2. Voltage change in the bidistillate for changing the distance of the measuring electrode from the cooled surface and the cooling time (cooling temperature $T = -30^{\circ}$);

1--for the distance of the measuring electrode of 30 mm; 2--20 mm; 3--10 mm.

Key: a--voltage, volts; b--hours.

It was established by tests that the lower the cooling temperature is and the more intense the loss of heat is through the cooled bottom, the greater the value of the originating voltage when cooling in a bidistillate and an NH₄Sl $(7 \times 10^{-5} N)$ solution. The maximum value of the voltage is reached faster with a lower cooling temperature.

Table 2

Relationship between maximum values of voltages and the intensity of cooling and heat loss through the cooling surface.

. 1	• • • • • • • • • • • • • • • • • • • •	· - S formerier mer				Pactoop NH _i Cl (7.50 + N)			
v	Сядра "мери и веря меть	Anches in	Herwit spirica use was instituted in the was in the control of the	Trad - 30' E. Hand foren	Buren diner. Inis arreits Ratherer in Treitinati	Tota 2 P C.,	Rucher date of the control of the co	Toxa do C.	Brain process
	Патунь нержавею- шая галь	1,3	60	-2,9	43			-2	80
	Патунь алючиний	-1,4		-7,2	26	1		7,4	25
	Латунь красная медь	3,0	84	-8,0	21	-4,6	89	- 13,0	!

Key: 1—cooled surface; 2—brass-stainless steel; 3—brass-aluminum; 4—brass-red copper; 5—bidistillate; 6—solution; 7— $T_c = -18^{\circ}C$, max. voltage, volts; 8—time for reacting max. temperature, minutes; 9— $T_c = -30^{\circ}C$, maximum voltage, volts; 10— $T_c = -20^{\circ}C$, voltage, volts; 11— $T_c = -30^{\circ}C$, max. voltage; 12—Note. 1. Thermal conductivity coefficient: brass-170 kilocalories/m. hour. degree; stainless steel-13; aluminum-175; red copper-300. 2. Distance between electrodes n = 10 mm.

Table 3

Change of maximum voltage in a bidistillate and a weak solution of NH₄OH depending upon the distance of the electrode from the cooled surface

l.		2 Inua	707.797	Paction NE. 7 3 2,5 10-4		
	Расстоя ине между электрода ин п. мм	Мансималь- мый потенци ал, «	Врия Финса ции макси- мального по- тенциала, мин	Мансичаль ный потенци- ал, в	Премя финса- пин мансы- мального по- тепциала, мых	
	30	-3.3	116	5.3	89	
	20	−7.8 .	97			
	12			13,5	58	
	10	-12	6)			

Key: 1--distance between electrodes h, mm; 2--bidistillate; 3--solution; 4--maximum voltage, volts; 5--time for reaching the max. voltage, minutes; 6--Note. The cooling temperature $T = -30^{\circ}C$

The curves of change in the voltage depending upon the distance from the electrode to the cooled surface and the cooling time are shown in curves (Fig. 2).

Tests established that the maximum voltage decreases with the greater distance of the measuring electrode from the cooling front, while the time of origination of the potential, counting from the start of cooling, increases.

The maximum voltage originating in opensided cooling of NH4OH solutions depends upon the concentration of the solution (Table 4).

Table 4

Relationship between maximum voltages of NH₄OH solution and its concentration

Ŋ	Концентра- ция раствора, N	Мянсималь вый потенци- ал 4 и его вран	Время фиксации мак- сима "БНОГО ПОТОНЦИ- вля от начала охлаж- дения, мин	() Концентрация раствора, М	Мансималь- ний потенци- ал в и его внак	Время финсации мансимального потенциала от начала оклаждения, мин
	10 - 10 - 4 5 - 10 - 6	-0,15 -0,33	106 82	1,5-10-	$ \begin{cases} -10 \\ -12 \\ -4 \end{cases} $	23 104
	3-10-	$\begin{cases} -18.3 \\ -23 \end{cases}$	{ 40 150	1.10-4	4	67

Key: 1--concentration of solution; 2--maximum voltage and its polarity, volts; 3--time for reading the maximum voltage from start of cooling, minutes; 4--Note. 1. Cooling temperature $T = -30^{\circ}C$. 2. Distance between electrodes h = 28 mm.

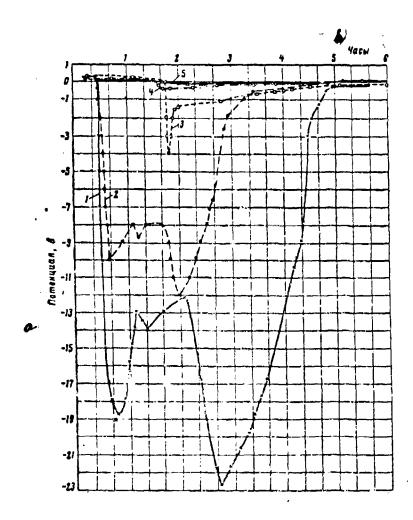


Fig. 3. Voltage change in NH₄OH solutions of various concentrations depending upon the cooling time (cooling time $T = -30^{\circ}C$; distance between electrodes h = 28 mm):
1--for concentration 3 x $10^{-5}N$; 2--1.5 x $10^{-5}N$; 3--1 x $10^{-6}N$; 4--5 x $10^{-5}N$; 5--10 x $10^{-5}N$.
Key: a--voltage, volts, b--hours.

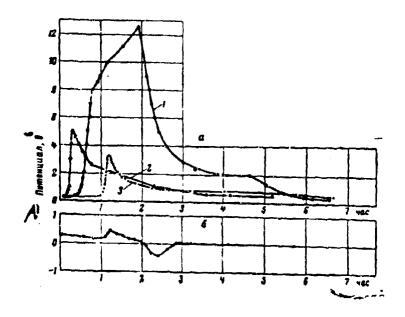


Fig. 4. Voltage changes in NaCl solutions of various concentrations depending upon the cooling time (T = -30°, h = 28 mm): a: 1--concentration of solution 3 x 10^{-3} N; 2--9.8 x 10^{-3} N;

 $3--16.5 \times 10^{-3}N$;

b: for concentration of 3×10^{-4} N;

Key: A--voltage, volts; B--hours.

Curves showing voltage changes in NH₄OH solutions of various concentrations depending upon the cooling time are shown in Fig. 3. The tests prove that the maximum voltage corresponds to a certain concentration of NH₄OH (maximum voltage was obtained at concentration 3 x 10^{-5} N).

The maximum voltage originating in the one-side cooling of weak solutions of NaCl depends on the concentration of the solution and the cooling temperature (Table 5).

Curves of the voltage change depending upon the concentration of the NaCl solution, temperature and cooling time are shown in Figs 4a and 4b. The maximum voltage corresponds to a certain concentration of the NaCl solution. The maximum voltage was obtained at a concentration of 3×10^{-3} N.

The maximum voltage increases with a higher cooling temperature. Tests of an NaCl solution with a $3 \times 10^{-3} N$ concentration were not taken into account in establishing this pattern because they were conducted with a not too stable cooling temperature.

Relationship between maximum voltages in the NaCl solution and its concentration and the cooling temperature

9		 Температура склаждения 						
		-	20*	-30°				
	Жонцентрация раствора, N	3) Мансималь- ямя пртенци- ал, е	ф) Время манси- мального по- тенцияла, лин	LOSSHIR.	У) Время макса- мельного по- тенциала, мин			
	3-10-4	{ +0,62 -0,49	{ 60 127	{+0,45 -0,38	69 140			
	3-10-●	{ 9,3 9,2	{170 * {260	12,5	110 **			
	9,8-10-2	8,1	28	5	15			
	18,5-10**	4,6	45	3	0.5			
	3-10-1	0,43						
1		чин — 20 15°; чин — 20 15°; чине — 20 40°.	•	ı	1			

Key: 1--concentration of solution; 2--cooling temperature; 3--maximum voltage, volts; 4--time to reach maximum voltage, minutes; 5--cooling; 6--Note. Distance between electrodes N = 28 mm.

The maximum potential originating in the solution depends on the composition of the dissolved salts. According to the tests made by the author and the investigations by E. Y. Workman and S. E. Reynolds (Table 1) a voltage originates in the NH₄OH solution that is about double that in the NH₄Cl solution. The great spread in the maximum voltages of solutions of NH₄OH (3 x 10⁻⁵N), NH₄Cl (7 x 10⁻⁵N) and NaCl (3 x 10⁻³N) (Tables 2, 4 and 5) and of maximum voltages of similar solutions with the same concentrations in tests by E. Y. Workman and S. E. Reynolds may be explained by the changes in the conditions of cooling intensity. The cooling of the solution in the tests done by the author occurred through a three-layer medium: brass-air-aluminum or copper, while in the Workman-Reynolds tests the cooling occurred through copper-mercury-copper; the removal of heat in that case was more intensive.

The reason for the difference in test results is confirmed by the dependence of the maximum voltages on the cooling temperature and on the heat loss through the cooled surface (Table 2). It should be noted that the spread in maximum voltages depending upon the cooling conditions

of NH4OH and NH4Cl solutions in these tests as compared to data by E. Y. Workman and S. E. Reynolds (Table 1) is approximately 10 times and in NaCl solutions only 2.4 times. Bidistillate tests observing simultaneously the time of origination of the voltages and the zero temperature on the inner surface of the copper indicated that the voltage originated before the zero temperature, at which ice may be formed, was established.

On the basis of the experimental data it may be assumed that the reason for voltages originating in cooling one side of the weak solutions is the tructural rearrangement of the water preceding freezing. Ground systems contain the usual ionic solutions, as well as suspensions. It is, therefore, expedient to clarify what voltages originate in freezing suspensions.

A solution of suspensions, if other dissolved ions are absent, differs from true suspensions in that clay particles in suspensions are charged negatively, while positively charged exchange cations are located on the surface of the particles or in the diffusion film, i.e., in the boundary phase.

The author made tests with suspensions of fractions $< 1 \, /\!\!\!\!/$ prepared from monomineral natural and monoionic kaolin and gumbrine clays. The basic material of gumbrine is montmorillonite. The saturation of kaolin and gumbrine with exchange cations of Fe'', Ca'', Na and NH4 was made in accordance with the method described in an article by Z. A. Nersesova (1961).

Results of the voltage measurements indicate (Table 6) that the mineralogical composition of clay parts, density of suspensions and the composition of exchange cations have considerable effect on the value and polarity of the maximum voltage. This effect is expressed in the following:

1. When cooling suspensions of equal density, the maximum originating voltage is greater in suspensions of kaolin saturated with Ca, Na or NH. A different relationship is observed for gumbrine and kaolin of natural composition. This may be explained by the various compositions of exchange cations on the surface of their particles or by a different content of soluble salts. Soluble salts were removed from Fe., Ca, Na and NH, gumbrines and kaolins in the process of preparing monoionic clays.

Table 6

Maximum voltages when cooling suspensions ($T = -30^{\circ}$) depending upon the density of suspensions, the mineralogical composition of clays and the composition of exchange cations.

Ŋ	Tanga	Ofmentue Hathoms	1 Плотность	Началь. Ими по-	Мансималь- име яначения	Время Финса-	7 Havant-
			суспена яй , е/А	тенциал,	потенциала при замерав- жин, с	Мального по- тенциала, муж	nepstypa cycnemasa °C
8	,	10) Естественный	∫ 1,424	0,115	6,2	63	20
_	×	Fe"	11,424	0,13	3,9	34	:•
	# d	Ca"	0,82	0,01	0,035	24	17
	9	GR.	0,54	0,05	0,95	27	18
	=	Na.	₹ 0,51	0,07	0,06		23
	-		1,54	0,045	0,03	·	25
	-	NH'₄	1 1,04	0,23	0,20	82	40
			1,04	0,18	0,18	105	16
Ą		Естественный	(1,204	0.04	, 70	40	
			1,204	-0,01 0,01	1,70	19	18
	=		10.04		1,30	20	••
	#	Fe"	0,04	0,102 0,12	-0,13	72	20
	14	Ca"	0,52	0,12	0,18	75	19
	°	Na.	(1,38	0,055	6,40	25	17,5
	×	• • • • • • • • • • • • • • • • • • • •			0,13	87	23
		NH4'	1,38	0,03	0,20	70	24
	- 1		{ 1,100 1,100	0,1	-2,20	20	17
			(1,100	0,2	←2,30	26	•
•	•	Естественный	0.200	0.44		19	
	E 0	Fe"	0,208	0,14	-0,70	23	22
	9	Ca"	0,208	0,02	-0,095		17
	×	Na.	0,208	0,065	-0,15	13 118	17
	7	NH.	0,208	0,05	-0,065		40
	_ {	AII.	0,208	0,04	0,15	161	16
4)	<u> </u>	Естественный	0,208	0.43	0.40	20	400
	5	NA'		0,12	0,19	22	17,5
	Kaonen	NH4	0,208	0,08	0,47	99	25
	-	47.748	0,208	0,05	~0,43	44	16

Key: 1--clay; 2--exchange cations; 3--density of suspensions, grams/ liter; 4--initial voltage, volts; 5--maximum voltage when freezing, volts; 6--time to reach max. voltage, volts; 7--initial suspension temperature; 8--gumbrine; 9--kaolin; 10--natural. 2. The polarity of the voltage at the measuring electrode in the solution depends on the suspension density. Thus, when the suspension density decreases the voltage polarity at the measuring electrode changes from positive to negative. This indicates that for a considerable density ($\rho \ge 0.52$ g/l) of suspensions the ice is charged negatively and the solution positively. At a small density ($\rho \le 0.208$ g/l) a reverse picture was observed: the ice is positive and the solution is negative.

Exceptions to this rule are suspensions made of kaolin saturated with exchange cation NH_4 in which a negative voltage originated with respect to the ice at a considerable suspension density ($\rho = 1.1$ g/1).

The obtained patterns in changing the maximum voltages and polarities when freezing suspensions saturated with cations Fe⁻⁻⁻, Ca⁻⁻, Na and NH₂ will be considered, comparing them with the origination of voltages when freezing the solvent (bidistillate). Maximum voltages of -13 or -17 volts were obtained in a bidistillate under test conditions identical to those in tests on freezing suspensions. In the paper by E. Y. Workman and S. E. Reynolds it was established that the origination of considerable voltages in the bidistillate was related to its content of ammonia which is a stable admixture of water.

A content in the bidistillate of suspended clay particles in a small amount (suspension density $\rho \leq 0.208$ g/l) reduces the voltage, but the polarity remains the same, i.e., ice is charged positively and the solution negatively. The same pattern remains for freezing suspensions prepared from kaolin ($\rho = 1.10$ g/l) saturated with exchange cations NH4, as well as in solutions of salts NH40H and NH4Cl. The absolute voltage at small suspension densities changes depending upon the mineralogical composition of clay particles, less in kaolin than in sumbrine suspensions.

When the number of suspended particles increases ($\rho \geqslant 0.52$ g/l) the polarity originating in freezing changes, i.e., ice is charged negatively and the solution positively. This same phenomenon originates when the solution contains small quantities of NaCl and CaCl salts.

At a high density of suspensions, the absolute value of voltage changes, depending upon the mineralogical composition of clay particles, more in kaolin than gumbrine suspensions, i.e., a pattern is observed which is the reverse of the pattern noted in suspensions with low density.

It is obvious that the patterns for voltage changes described above for solutions (salts and suspensions) must take place also in ground solutions.

It is well-known that when drying grounds by means of electrical current, the water in the ground migrates from the positively charged electrode to the negatively charged electrode. In tests with salt solutions done by E. Y. Workman and S. E. Reynolds (1950) and in the tests made by the author not only on salt solutions, but also on suspensions, a change of polarity was observed on the electrode located in the solution. It may be assumed on the basis of this data that the extent of moisture migration in the ground, along with previously known factors, may also be affected to a considerable extent by the voltages originating in ground solutions on freezing. The degree of the voltage effect on migration will depend on the concentration of salts in ground solutions, on the mineralogical composition of colloidal particles in the suspension solutions, and the composition of exchange cations on their surface.

Drying ground by means of electrical current depends not only on the value of the voltage applied to the electrodes, but also on the time of its action and the distance between the electrodes. Therefore, the obtained dependence of the originating voltage on the temperature and cooling intensity was considered, taking all three factors into consideration. The author measured originating voltages at a distance of 10 mm.

Obviously, the migration of moisture in the ground does not require considerable voltage. In this case, the value of the migration to the freezing front is determined not so much by the value of the voltage originating in the solutions in the ground, as by the time of its action.

In considering the effect of voltages originating in the ground on the migration of moisture, the action of other factors considered earlier should not be neglected. It may be assumed that several factors may be effective at the same time but in a certain sequence. The author assumes that the primary reason for the migration of moisture to the freezing front is the voltages originating in the solutions in the ground that cause a movement of the moisture from the near layer to the freezing front and create a scarcity of moisture in this layer which is replenished by a flow of moisture from the lower layers under the effect of the moisture gradient.

The interrelationship between these factors, affecting the movement of moisture, will determine the value of water migration (or the value of swelling).

At this stage of studying the question of ground swelling it is necessary to take into account not only the composition and moisture content of ground and the distribution of ground waters, but also to have information on the composition and concentration of ions in the ground solution at the start of freezing.

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